RADIO CORPORATION OF AMERICA
RCA LABORATORIES

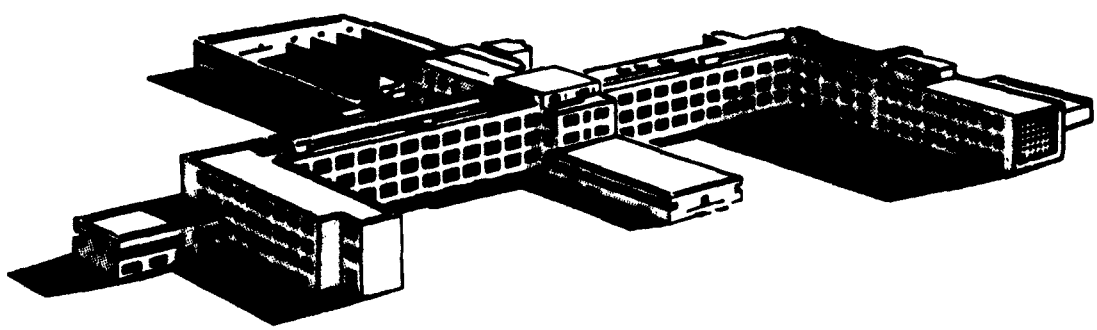
MICROWAVE AND OPTICAL MASERS FOR MM WAVES

REPORT NO. 1

CONTRACT NO. DA36-039-AMC-00082 (E)

FIRST QUARTERLY REPORT
FOR THE PERIOD
NOVEMBER 1, 1962 TO JANUARY 31, 1963

U. S. ARMY ELECTRONICS RESEARCH AND DEVELOPMENT LABORATORY
FORT MONMOUTH, NEW JERSEY



DAVID SARNOFF RESEARCH CENTER
PRINCETON, NEW JERSEY

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OBJECT

To investigate the feasibility of optically pumped coherent far infrared (5μ – 1000μ) radiation generators.

Prepared by: Z. J. KISS

Approved by: H. R. LEWIS

RADIO CORPORATION OF AMERICA
RCA LABORATORIES
PRINCETON, NEW JERSEY

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PURPOSE

The purpose of this contract is to construct coherent far infrared ($5\mu - 1000\mu$) radiation generators. To fulfill this aim, solid and gaseous media suitable for maser action will be investigated. Instrumentation to study the far infrared region of the spectrum will be completed. A study of the lattice spectra of solid maser hosts will be carried out and the energy levels of impurities in the various hosts will be determined.

ABSTRACT

In this report a partial account is given of the instrumentation required for the study of optical properties of possible far infrared (5μ – 1000μ) coherent radiation generators. The operation of a laboratory-built Michaelson interferometer is described together with a Perkin-Elmer No. 301 spectrometer and the associated cryogenic equipment. Preliminary study of the lattice absorption spectrum of $CaWO_4$, $CaMoO_4$ and $PbMoO_4$ is presented. A possible 5μ optical maser transition in the $CaWO_4:Nd^{3+}$ system is evaluated.

PUBLICATIONS, LECTURES, REPORTS, AND CONFERENCES

During the report period no publications or reports were issued, and no lectures or conferences were attended.

FACTUAL DATA

I. INTRODUCTION

Since the announcement of the first optical maser[1] in 1960 maser action has been achieved in a large number of systems employing insulating solids, gases, glasses, semiconductors, and organic chelates as the medium. The longest infrared maser frequency was reported[2] at 18.8μ using a neon gas system. Microwave masers on the long wave length end of the spectrum have been operated since 1954, and the highest frequency microwave maser reported to date[3] is at 75 kMc using iron-doped rutile as the active medium. Between these two regions, however, lies a large gap, most of the "far infrared" (from 5μ to 1000μ) where until now there are no oscillators available. This situation is particularly inadequate since the very weak thermal sources combined with insensitive detectors make this region of the spectrum very difficult to study.

The object of the present contract is to investigate the optical properties of materials (solids and gases) in the far infrared region of the spectrum to find suitable systems for the construction of maser oscillators.

The present report describes the effort of the first three months that was expended in three areas:

1. The construction of the instrumentation necessary for the study of the far infrared. This involves the building of a Michaelson interferometer spectrometer to be used in the 150μ to 1000μ region, setting up a Perkin-Elmer 301 double-beam-grating spectrometer to be used in the 15μ to 200μ region, and assembly of cryogenic equipment suitable for optical studies in the above regions of the spectrum.
2. A near infrared study of some of the insulating solid host lattices that will be used in the maser material search.
3. A study of a possible maser system using the Nd^{3+} as the active ion with expected maser output at 5μ.

II. EXPERIMENTAL DETAILS

A. MICHAELSON INTERFEROMETER SPECTROMETER

Spectroscopy beyond 5μ could be called "intensity-limited spectroscopy" since the limit of spectral resolution is not determined by the dispersing elements (prisms or gratings), but by the available source intensity. In such cases a spectrograph is preferable to a spectrometer. Where there are no photographic films available, the same gain in integrating time over a scanning spectrometer can be achieved with a Michaelson interferometer spectrometer.

The Michaelson interferometry method which allows simultaneous observation of all parts of the spectrum is called the Fourier transform method. The method has two basic advantages. First, the instrument has cylindrical symmetry, and circular slits can be used with more efficient use of the optical elements; but more important, comparing the interferometer to a scanning device, both with the same total observation time T, if the scanning spectrometer covered M spectral elements in time T, there would be a gain in the signal-to-noise ratio of $\sqrt{\frac{T}{T/M}} = \sqrt{M}$ using the Fourier transform method.

A schematic diagram of the Michaelson interferometer that we have constructed is shown in Fig. 1. A parallel beam of light from a source S of spectral distribution $B(\nu)$ is divided at the

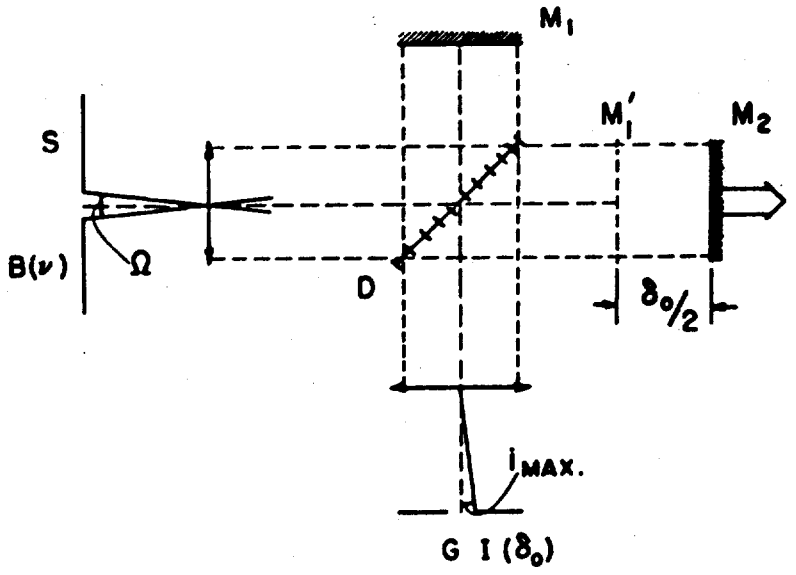

Fig. 1. Schematic diagram of a Michaelson interferometer spectrometer. (S is the source of brightness $B(\nu)$, D is the semireflecting surface, M_1 is the fixed mirror, M_2 is the moving mirror, M_1' is the image of M_1 in D, δ_o is the path difference and G is the detector.)

semireflecting surface D into two parts. These are reflected at the plane mirrors M_1 and M_2, respectively, returned to D and recombined at the surface of D after having traversed paths differing by an amount δ. The intensity $I(\delta_o)$ falling on the detector G is recorded as the mirror M_2 is moved uniformly; that is, the path-difference δ_o is changed linearly with time. If we consider a monochromatic point source $B(\nu_o)$, the interferogram will be

$$I(\delta_o) = \frac{B(\nu_o)}{4} \quad (1 + \cos 2\pi\nu_o\delta_o) \tag{1}$$

That is, a regular cosine function as a result of the constructive and destructive interference between the two beams as the path difference δ_o is varied. Or for an arbitrary frequency distribution $B(\nu)$ the δ_o dependent part of the interferogram

$$r(\delta_o) = 1/4 \int B(\nu) \cos 2\pi\nu\delta_o \, d\nu = T\,[B(\nu)] \tag{2}$$

which is the Fourier cosine transform of the spectrum under study. The Fourier method of spectroscopy using a Michaelson interferometer then consists of recording the interferogram $r(\delta_o)$ and performing a Fourier transformation using computers to obtain the intensity distribution $B(\nu)$ of the frequency spectrum under investigation.

The detailed theory of the Fourier transform method of spectroscopy has been worked out in great detail.[4-7] Only the two major factors will be outlined here that determine the limit of resolution of the method. The first difficulty stems from the fact that the source has a finite extent Ω, and while the path difference for the axial ray is δ_o, the path difference for the rays at an angle i (Fig. 1) is different. If we assume that there is a distribution of path differences $D(\delta_i, \delta_o)$, then the actual recorded interferogram will not be $r(\delta_o)$ as given in Eq. (2) but another function $A(\delta_o)$ where

$$A(\delta_o) = \text{constant} \int D(\delta_i, \delta_o)\, r(\delta_i) \, d\delta_i = r(\delta_o) \ast D'(\delta_o). \tag{3}$$

While the effect of Eq. (3) can be minimized by using sufficiently small slits, there is a more fundamental limitation on the resolution. Namely, the interferogram is not defined for all values $-\infty < \delta_o < \infty$, but only in a limited range $-L < \delta_o < L$ where L is the maximum path difference of scan. That is, the final interferogram is $I(\delta_o) = A(\delta_o)\, E(\delta_o)$ where $E(\delta_o)$ is a delta function of unit height in the interval from $-L$ to $+L$. Therefore, the Fourier inverse of $I(\delta_o)$ will not give the true $B(\nu)$ but a resolution-limited $B'(\nu)$

$$B'(\nu) = T\,[I(\delta_o)] = T\,[r(\delta_o)]\; T\,[D'(\delta_o)]\; T\,[E(\delta_o)] \tag{4}$$

Neglecting for the moment the effect of the extended slit ($T\,[D(\delta_o)] = 1$), the calculated spectrum is the convolution of the true spectrum, $B(\nu)$, with the Fourier transform, $f(\nu)$ of $E(\delta_o)$. The function $f(\nu)$ is given by the equation

$$f(\nu) = 2L\,\frac{\sin 2\pi\nu L}{2\pi\nu L} \tag{5}$$

and is called the instrumental line shape function (Fig. 2). The true spectrum $B(\nu)$ is scanned by the instrumental line shape function in analogy to the way that a grating spectrometer scans the

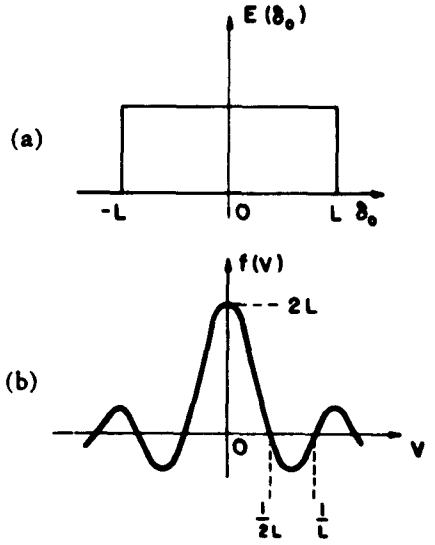

Fig. 2. a. The natural weighting function due to limited travel L of mirror M_2.
b. The natural instrumental line shape junction.

spectrum with a particular line shape. The limit of resolution in terms of maximum travel can also be deduced from Fig. 2. For a point source using Rayleigh's criterion (two frequencies separated by $\Delta\nu$ can be resolved if the maximum of one in the Fourier pattern of Fig. 2 coincides with the minimum of the other) the limit of resolution is $\Delta\nu = 3/8L$ when an extended source is used, using a slit that gives optimum compromise between luminosity and resolution[7] $\left(\frac{r_o i_m^2}{2} = \frac{1}{L}\right)$ the actual limit of resolution is $\Delta\nu = 1/L$.

Figure 3 shows a photograph of the actual spectrometer that we constructed. The collimating system uses 3-in. diameter $f/4$ off axis parabolic mirrors, and mirror M_2 can be scanned 10 cm

Fig. 3. Photograph of Michaelson interferometer. The collimating system employs 3-in. aperture / 4 off axis paraboloids.

(L 20 cm) giving a limit of resolution of 0.05 cm^{-1}. The instrument was tested using semisilvered quartz beam splitters and a continuously operating $CaF_2:Dy^{2+}$ maser at 2.36μ as the source. The interferogram is shown on Fig. 4. Work is in progress to enclose the interferometer in a vacuum

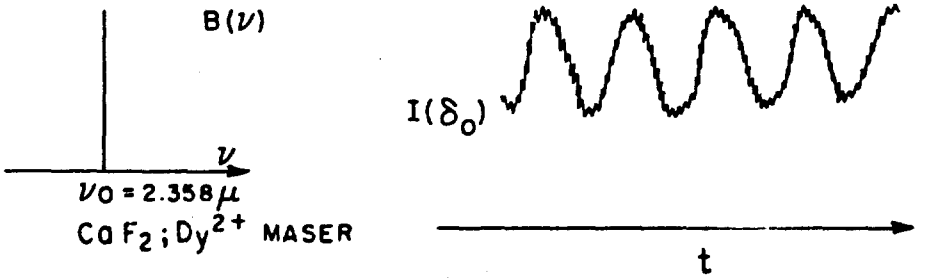

Fig. 4. Interferogram of a 2.36μ $CaF_2:Dy^{2+}$ optical maser line.

chamber and to extend the operation to the far infrared range of 100μ to 1000μ using stretched mylar film as a beam splitter. Details of the numerical Fourier transformation and the experimental evaluation of the instrument function will be given in the next quarterly report.

B. PERKIN-ELMER NO. 301 GRATING SPECTROMETER

A Perkin-Elmer No. 301 double-beam-grating spectrometer was installed and operated covering the range from 15μ to 200μ. A schematic diagram of the instrument is shown in Fig. 5. To cover the

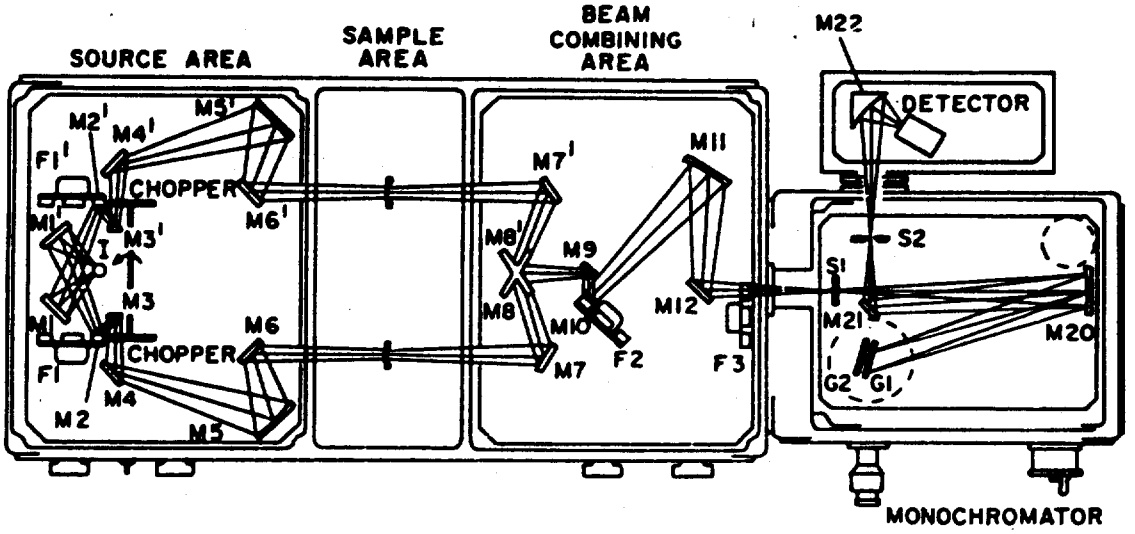

Fig. 5. Schematic diagram of the Perkin-Elmer No. 301 grating spectrometer.

above region the instrument uses four interchangeable gratings ranging from 40 lines/mm to 8 lines/mm. We believe that with the use of a 4 lines/mm grating the useful range of the spectrometer can be extended to 300μ. The spectrometer can be used in both single- and double-beam operation for reflection absorption and emission spectra studies and the intensity-limited resolution is about 1 cm^{-1}. Examples of spectra taken on this instrument are shown in Figs. 6, 7, 8 and 10.

C. CRYOGENIC EQUIPMENT

A liquid helium dewar has been constructed for the use in the far infrared optical studies. Provisions are made both for cold finger contact cooling and for immersing the specimen in the liquid coolant. The room temperature vacuum seal of the NaCl, CsBr, quartz and mylar windows is achieved by using rubber and Teflon O-ring seals. The sealing of the windows at helium temperature is done in two steps: 1. By using indium and gold O-rings for the quartz windows.. 2. The other window materials are araldited to a 3-mil thick copper ring that takes up the difference in contraction between the window and the metal dewar, and the copper ring is soft-soldered to the metal body.

III. LATTICE ABSORPTION EDGE OF MASER HOST MATERIALS

In the construction of far infrared solid-state masers the greatest difficulty will be to find host materials with windows in the lattice absorption bands where the host absorption losses are not too great to prevent maser action. Since detailed study of the lattice spectrum for most solid-state maser hosts is not available we have started a compilation of the infrared absorption and reflection spectra of single crystals of these hosts.

Figure 6 shows the near infrared absorption edge of $CaWO_4$ as a function of temperature. It is interesting to note that after the absorption band at 5μ there is a transmission window at 10μ. We believe that the 5μ absorption band corresponds to a molecular-like vibrational spectrum due to the vibrational modes of the WO_4 complex. The $CaWO_4$ belongs to the $C_{3h}{}^6$ point group. In the structure the Ca atoms have a large bond distance to the WO_4 and it is reasonable to assume

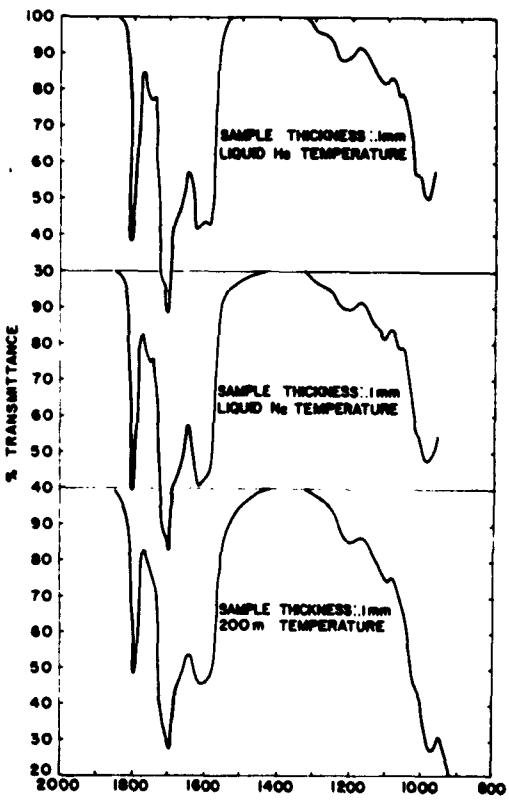

Fig. 6. The near infrared absorption spectrum of $CaWO_4$ at different temperatures.

that the WO_4 complex can be considered as a fairly isolated system. The local symmetry of the WO_4 group is S_4 and the $3N - 6 = 9$ modes of vibration are expected to break down into A and $E + 2T_2$ vibrations. Of these vibrations the E and one of the T_2 are expected to be infrared active. At low temperatures one can detect considerable structure on the 5μ absorption band of Fig. 6.

The different components may be due to the second, third or fourth overtones of the infrared active vibrational modes. A more affirmative identification will have to await more quantitative calculations of the vibrational pattern. The host absorption of $CaMoO_4$ and $PbMoO_4$ in Fig. 7 is very similar to that of the $CaWO_4$. This is not surprising since the MoO_4 complex is very similar in all aspects to the WO_4, including the reduced mass.

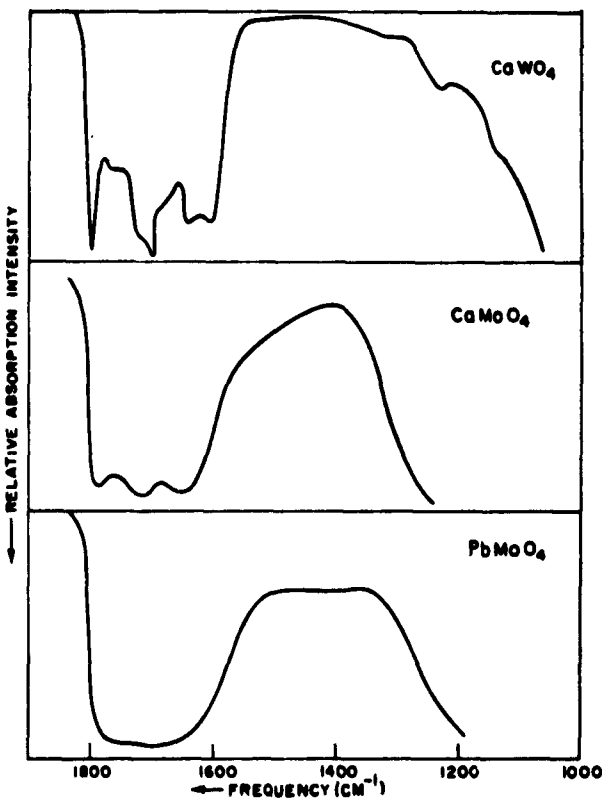

Fig. 7. The near infrared absorption spectra of $CaWO_4$, $CaMoO_4$, and $PbMoO_4$ at liquid nitrogen temperature.

A reflection spectra of the *reststrablen* frequencies of the $CaWO_4$ host is shown in Fig. 8 in the $20\mu \rightarrow 50\mu$ range. An analysis of the lattice modes will be given in the next quarterly report.

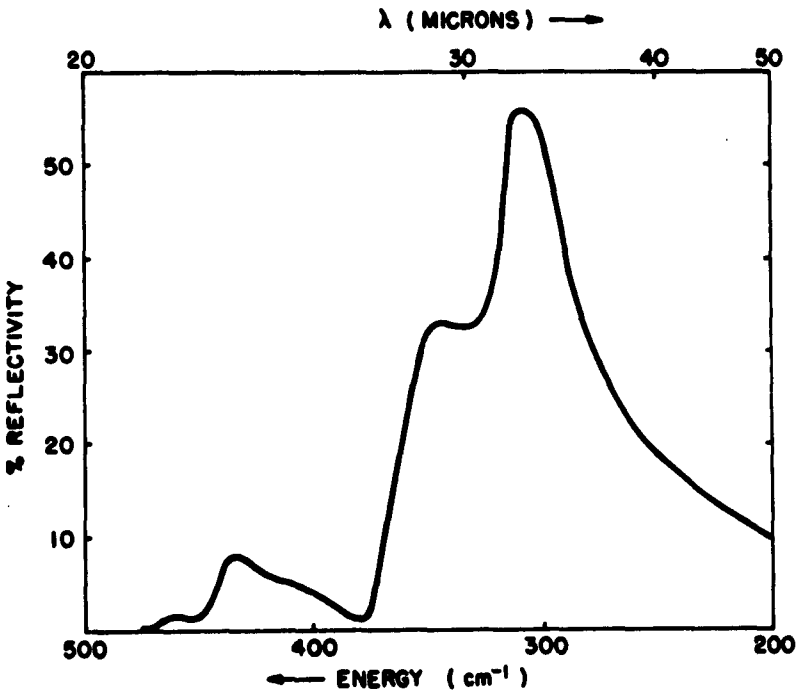

Fig. 8. The *reststrablen* reflection spectra of $CaWO_4$ in the 20μ to 50μ range.

IV. THE 5μ MASER TRANSITION IN Nd^{3+}

One of the most suitable active ions for solid-state maser systems is the Nd^{3+} ion. The lowest spin orbit multiplet is the ^4I multiplet (Fig. 9), and the optical maser transition is between the $^4F_{3/2}$ level and the $^4I_{11/2}$ levels of the lowest multiplet. This transition occurs at the

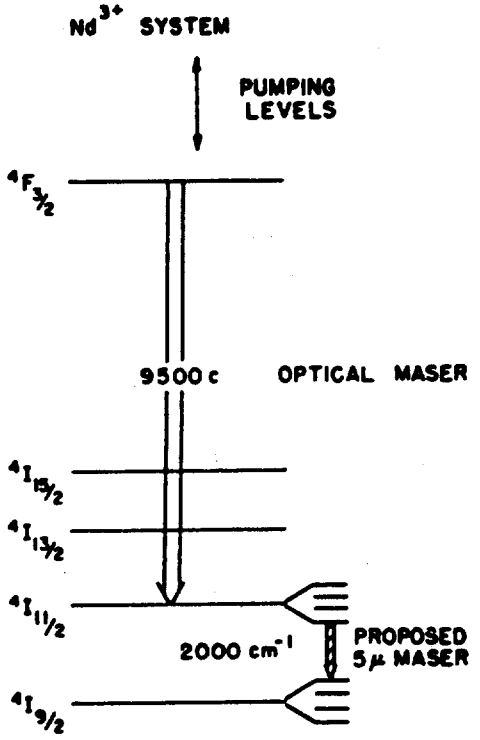

Fig. 9. Energy levels of the Nd^{3+} system in solid hosts.

9500 cm^{-1} region (1.06μ) and terminates 2000 cm^{-1} above ground state on the $^4I_{11/2}$ level. There exists a possibility for further four-level maser action between the $^4I_{11/2}$ state and a crystal field split component of the ground $^4I_{9/2}$ state. The fluorescence spectrum of $CaWO_4$:Nd^{3+} is shown in Fig. 10a. Emission from the $^4F_{3/2}$ level to the lowest three components of the ^4I spin-orbit multiplet can be seen. The absorption spectrum corresponding to the $^4I_{9/2}$ to $^4I_{11/2}$ transition can be seen in Fig. 10b. The *4I-4I* absorption just misses the $CaWO_4$ absorption at 5μ, and since the resonance transition is at 2000 cm^{-1}, part of the possible $^4I_{11/2}$ to $^4I_{9/2}$ emission terminating

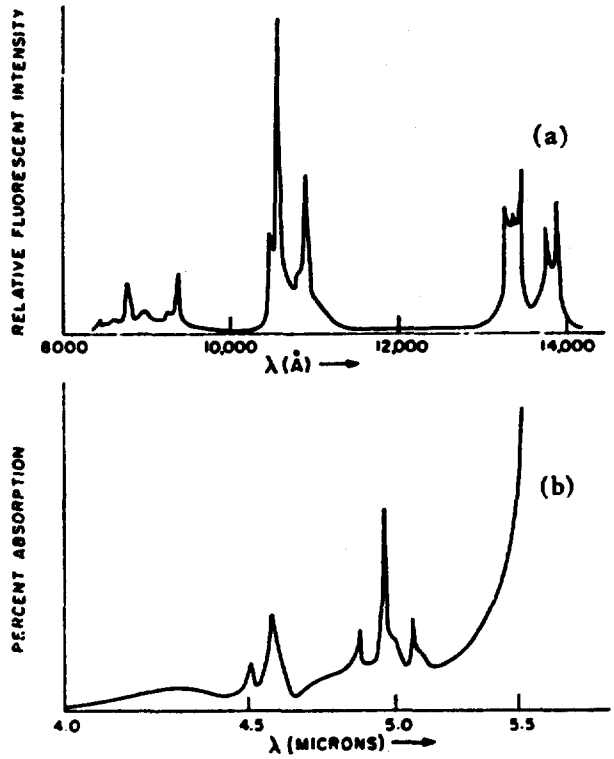

Fig. 10. a. The fluorescence spectrum of $CaWO_4:Nd^{3+}$ at liquid nitrogen temperature.
b. The absorption spectrum of $CaWO_4:Nd^{3+}$ corresponding to the $^4I_{9/2} \cdot {}^4I_{11/2}$ transition.

on the crystal field split components of the $^4I_{9/2}$ state would be buried in this $CaWO_4$ absorption. Efforts to observe emission in the $CaWO_4:Nd^{3+}$ system at 2000 cm^{-1} were unsuccessful. This is not too surprising since due to the nearness of the lattice absorption the nonradiative decay of the $^4I_{11/2}$ state must be very fast. A further attempt to observe emission will still be made while the $^4I_{11/2}$ state is pumped by the optical maser transition at liquid helium temperature. Furthermore, even if this transition will not prove suitable in the $CaWO_4$ host, the Nd^{3+} system will have to be tried in other hosts, where the lattice absorption edge is further out in the infrared.

CONCLUSIONS

The performance of the laboratory-constructed Michaelson interferometer spectrometer was evaluated and was found satisfactory. A Perkin-Elmer No. 301 spectrometer was installed and tested by studying the absorption spectra and reflection spectra of solid host materials for masers. The possibilities of a 5μ maser transition in the $CaWO_4:Nd^{3+}$ system were evaluated.

PROGRAM FOR NEXT INTERVAL

1. Extend the operation of the Michaelson interferometer spectrometer to the far infrared region of the spectrum.
2. Determine the near infrared lattice absorption edge of the alkaline earth halide and mixed halide systems and continue the measurements of the *reststrahlen* spectra of maser hosts.
3. Attempt maser operation in the $^4I_{11/2} \rightarrow {}^4I_{9/2}$ transition of Nd^{3+} in various hosts.
4. When the far-infrared instrumentation is completed, search for maser action in suitable systems starting with $CaF_2:Dy^{2+}$ system at 24 cm^{-1}.

IDENTIFICATION OF KEY PERSONNEL

The following members of the technical staff have contributed to the contract during the past work period:

A. Akselrad, Member of Technical Staff	480 hours
Z. J. Kiss, Project Engineer	200 hours

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